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GAS HOLDUP IN BUBBLE COLUMNS - A LITERATURE REVIEW

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GAS HOLDUP IN BUBBLE COLUMNS - A LITERATURE REVIEW

by

T.J.W. de Bruijn*

SUMMARY

Extensive data are available on the hydrodynamics of bubble columns. Unfortunately, published correlations do not agree with each other very well and do not predict the observed behaviour of large scale industrial reactors. To alleviate the discrepancies and to develop general correlations a research program was started with the University of Waterloo to study the dynamics of three-phase bubble columns on model systems. A review of the relevant literature on gas holdup is presented and a comparison is made of the predictions of four literature correlations for three air/liquid systems.

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RÉTENSION DE GAZ DANS LES COLONNES DE DISTILLATION - COMPTE RENDU DE LA LITTÉRATURE

par

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SOMMAIRE

Des données exhaustives sur l'hydrodynamique des colonnes de distillation sont disponibles; toutefois les corrélations qui ont été publiées ne correspondent pas parfaitement entre elles et ne prédisent aucunement le comportement observé dans les réacteurs industriels à grande échelle. Afin de palier aux contradictions et de développer des corrélations générales, on a créé un programme de recherche avec l'Université de Waterloo dans le but d'étudier la dynamique des colonnes de distillation à trois phases sur des modèles de système. Ce rapport présente donc un compte rendu de la littérature pertinente à la rétention de gaz ainsi qu'une comparaison faite entre les prédictions provenant de quatre corrélations pour trois systèmes air/liquide tirées de la littérature.

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INTRODUCTION

Bubble columns are a type of reactor being used in a variety of industrial processes, like, e.g., Fischer-Tropsch, hydrotreating, coal liquefaction, bioprocesses and hydrocracking. The CANMET hydrocracking process employs a three-phase co-current bubble column to convert heavy oils, bitumen and atmospheric and vacuum resids to lighter products.

The dynamic behaviour of bubble columns, of which gas holdup is only one aspect, is quite important since it influences the conversions that can be obtained, the solids accumulation and concentration profiles along the reactor and temperature control. Gas holdup specifically influences the effective liquid volume present in the reactor and thus directly influences the conversions or yields.

Despite extensive studies of gas holdup, liquid mixing or slurry mixing, recent results reported from large-scale reactors indicate that existing published correlations do not predict well the observed behaviour. In addition, published correlations predict different values even though all were developed from results obtained with laboratory equipment. To gain more insight into the variables causing these discrepancies and to develop more general correlations a research program was started with the University of Waterloo on model systems.

As part of this program a review of the relevant literature on gas holdup in bubble columns was made and is summarized in this report. Only low pressure, low temperature systems were considered. Also, as an example of the differences in gas holdup predictions, four literature correlations were compared for three different air/liquid systems, viz., air/water, air/trichloroethylene and air/Varsol (Varsol DX 3641 is a light hydrocarbon oil available from ESSO Chemicals).

REVIEW

Gas holdup is defined as the volume percentage of gas in the two- or three-phase systems under consideration. It can be measured in different ways, e.g., by the difference in manometer readings [Bhaga et al. (1971), Hikita and Kikukawa (1974), Hikita et al. (1980), etc.] by measuring the liquid height with and without gas flow (batch) [Akita and Yoshida (1973),

Hammer and Deckert (1978), Deckwer et al. (1980), Gopal and Sharma (1983), Oels et al. (1978), Patil et al. (1984), etc.] or by the use of different probes [Buchholz et al. (1983), Nottenkamper et al. (1983), Linneweber and Blass (1983), etc.]. The volume expansion and manometric pressure method compared relatively well (Nottenkamper et al. (1983), Kara et al. (1982)]. Optical probe measurements were about 10% lower than manometric pressure results [Nottenkamper et al. (1983)].

Despite the good agreement between various measuring methods, quite different correlations describing gas holdup are presented in the literature. Shah et al. (1982) list most of the important correlations, some of which are also listed in Table 1. Before the different literature correlations are compared, some factors influencing gas holdup are discussed briefly.

In principle, gas holdup depends on the bubble rise velocity and the number of bubbles. The bubble rise velocity is dependent on the bubble size, the liquid and gas properties (system properties) and liquid circulation patterns. Different equations have been applied to describe the bubble rise velocity.

Bubble size or bubble formation dynamics are very complicated and various models exist to describe bubble sizes for different gas rates and different mechanisms of formation [Wallis (1969) and Mersmann (1978)]. For example, the model for single bubble formation predicts the bubble size to be constant and dependent only on orifice size and system properties. At higher gas rates, the bubble size depends on the gas flow rate through the orifice. At still higher gas rates the momentum flux from the orifice becomes important and also the orifice area appears in the correlation describing bubble volume [Wallis (1969)]. At sufficiently high gas rates the bubbles are not formed individually but the gas leaves the orifice as a jet which breaks up into individual bubbles. The bubble size is then about twice the size of the orifice [Wallis (1969)].

Kumar et al. (1976) and Bhavaragu et al. (1978) report correlations for the bubble size containing orifice dimensions. Also, Voigt and Schugerl (1979), Kato and Nishiwaki (1972), Zuber and Hench (1962) and Bach and Pilhofer (1978) observed an influence of the orifice size. However, Akita and Yoshida (1974), Quicker and Deckwer (1981) and Patil et al. (1984) did not.

Another approach to predicting bubble sizes in bubble columns is to relate the bubble size to the power input. Bubble columns at normal operating conditions have a high degree of turbulence and bubble size might be determined to a large extent by shear stresses. For example, Hinze (1955) proposed the following formula to predict the bubble size:

$$d_B = 0.725\left(\frac{\sigma}{\rho_f}\right)^{0.6}\left(\frac{P}{M}\right)^{-0.4}$$

This approach has been taken by several authors: Voigt and Schugerl (1979) show a plot of the mass transport coefficient from gas to liquid, k_La, versus specific power input. The mass transport coefficient increases with increasing power input, however, the increase was found to be different for each perforated plate employed. It was explained by a difference in the fraction of microturbulence as compared to large scale turbulence. Perforated plates with small diameter holes disperse the gas more effectively than plates with large diameter holes [see also Buchholz et al. (1983)]. Quicker and Deckwer (1981) also found that higher energy dissipation (in this case from a single nozzle) resulted in a larger gas holdup for Vestowax. Buchholz et al. (1983) correlated the interfacial area with the energy input and explained that its effect depended on the type of medium, i.e., whether it was coalescing, non-coalescing or in-between. Finally, Oels et al. (1978) found that, in coalescence-hindering media, aerators with the highest local energy dissipation rate gave the smallest bubbles and largest gas holdup and k_La values. In coalescence promoting media, the aerator type did not influence the gas holdup or k_La. In general, a relation such as that proposed by Hinze is valid for non-coalescing media in which the size of the bubble formed at the sparger is larger than the bubble size in the bulk fluid. In coalescing media, the bubble size can be estimated from relationships such as those given by Heijnen and van't Riet (1984).

Coalescence and bubble breakup appear to be extremely important and greatly affect the gas holdup and k_La values. Whether or not the size of a primary bubble will change depends on its size relative to the maximum equilibrium bubble size and whether the system is coalescing or non-coalescing. Every system has a maximum equilibrium bubble size which depends on the surface tension and fluid stresses. For single particles, Mersmann (1978) equates the surface tension force to the buoyancy force and derives:

$$d_B = 2.45 \left(\frac{\sigma}{\Delta\rho g}\right)^{0.5}$$

for low viscosity liquids. If the bubbles formed at the sparger are already at or above their maximum equilibrium size, other components present will have very little effect [Oels et al. (1978), Voigt and Schugerl (1979)]. However, if the primary bubbles are smaller, the presence of other components affecting the coalescence rate, can result in large differences, the extent depending on the "strength" of the coalescence inhibitor [Buchholz et al. (1983)] and the liquid height, i.e., the bubble residence time [Voigt and Schugerl (1979)]. Large increases in gas holdup and k_La can result when coalescence inhibitors are used. Bubbles do not reach their maximum equilibrium size in such systems. Sparger design may greatly affect gas holdup and k_La under those circumstances since they "dictate" the primary bubble size.

Coalescence is influenced by surfactants, contaminants, and liquid properties, in fact by any substance altering the surface or interfacial properties of the gas bubble and the liquid. Therefore, mixtures can also show different behaviour than that of the pure liquids [Bhaga et al. (1971), Bach and Pilhofer (1978), Hammer and Deckert (1978), Godbole et al. (1984)]. For example, Bhaga et al. (1971) measured the gas holdup in different organic mixtures and found that for some mixtures it increased with an increase of the concentration of the second component, went through a maximum and then decreased. For other mixtures the gas holdup was constant over the whole range of compositions. They suggested that the increase in gas holdup is related to the difference in surface tension of the pure components. Also Bach and Pilhofer (1978) found existing correlations inadequate to describe the gas holdup in mixtures of glycerol and water, and in sodium sulphite solutions. Godbole et al. (1984) attributed the difference in holdup for alcohol solutions as compared to Soltrol-130 to an increase in surface activity of the bubbles and not to the bulk properties of the solution.

For mixtures of liquids which have significant differences in one or more physical properties, it appears that only those holdup correlations that were derived by using measurements on the mixture should be used, at least until such time as a better understanding of bubble dynamics and coalescence phenomena is available.

The above review illustrates the very complicated nature of bubble formation and breakup and the many factors that influence these phenomena. For engineering purposes a simple correlation (empirical or theoretical) is needed to describe the gas holdup directly as a function of the superficial gas velocity and of the important system properties. Use of correlations describing bubble sizes and bubble rise velocities to derive the gas holdup tends to be too inconvenient or imprecise for most practical purposes, given our present state of knowledge, and fails to provide greater accuracy. Therefore, emphasis is placed on correlations directly relating gas holdup with the superficial gas velocity and the physical properties of the system.

Typical literature correlations such as those given by Shah et al. (1982) contain some variables such as liquid density, surface tension and liquid velocity, implying that the correlations are suitable for describing all types of systems. However, Quicker and Deckwer (1981) noted that the existing literature correlations did not describe their measurements on xylene, decaline, C_{10}-C_{14} paraffins or Vestowax. Ying et al. (1981) found that Hikita and Kikukawa's correlation (1974) did not fit air/methanol data while Akita and Yoshida's correlation (1973) did. However, Ying et al. (1981) found that Akita and Yoshida's correlation failed to describe the tetralin/nitrogen data. Even for the most used system, air/water, fairly large differences in predicted gas holdup result when using different correlations. As discussed above, factors such as impurities, type of sparger and orifice size can have a pronounced effect of gas holdup particularly at low gas velocities, and the influence of these factors is not considered in most of the correlations. It appears that in order to ignore these factors, the primary bubble size must be equal to the equilibrium bubble size or the medium must be coalescence promoting, and the bubble column must have a liquid height such that the stable bubble size is reached in a fraction of the total liquid height. When choosing a correlation to describe a process it is important to check whether these conditions were met during the experiments which were used to derive the correlation.

Most correlations show a difference in their dependence of the gas holdup on the superficial gas velocity. It is well known that if the gas holdup is expressed as $\epsilon_G \propto U_G^n$, n depends on the flow regime. In the bubble flow regime, n appears to be close to 1, whereas at higher U_G (turbulent-churn regime) n becomes smaller [Deckwer et al. (1980), Bach and Pilhofer (1978),

Shah et al. (1982)]. Therefore, if investigators measure in both regimes and use all measured values to derive a single correlation, the value of n will be some average number which will depend on the number of measurements in each regime and the range of measurements in each regime, e.g., whether measurements were done up to 0.1 m/s or 0.20 m/s.

In addition to the factors mentioned above, the gas holdup may be influenced by a number of other variables which are not always present in the existing correlations such as column size, liquid height, liquid flow rate, gas density and the presence of solids.

COLUMN SIZE: Hughmark (1967) found that a column size of up to 0.10 m diam had an effect on gas holdup. Deckwer et al. (1980) also found a difference in holdup in a 0.041 and 0.10 m column. Hikita et al. (1980) measured holdup in a 0.10 m column and compared their results with those reported in the literature for columns larger than 0.10 m and found no appreciable effect. Nottenkamper et al. (1983) measured the gas holdup in 0.19, 0.45 and 1.0 m columns and obtained comparable results for the 0.19 and 0.45 m columns but lower holdup values for the 1 m column at high gas rates, which they attributed to the larger diameter. Gopal and Sharma (1983) measured the gas holdup in 0.2, 0.6 and 1.0 m columns and state that the column diameter did not influence the gas holdup. However, their results do show a difference in gas holdup values for the different column sizes, which could be due to the differences in column size or in spargers used. Ying et al. (1981) found no difference in gas holdup values in a 0.125 and 0.30 m column. Koide et al. (1979) measured the gas holdup in a 5.5 m column and found no significant difference with literature values reported for columns >0.10 m. Kato and Nishiwaki (1972) carried out measurements in 0.066, 0.122 and 0.214 m columns and found that the gas holdup increased with decreasing column size. Koide et al. (1984) observed smaller gas holdups and $k_L a$ values in columns smaller than 0.2 m.

Despite some contradictory results in the literature it appears that most investigators consider a 0.10-0.30 m column large enough to obtain gas holdup values which can be reliably used to predict holdup values in larger columns.

LIQUID HEIGHT: The liquid height influences the average pressure in the column and therefore the average superficial gas velocity. Based on the

selection of the vertical reference point, different superficial gas velocities will be calculated for the same mass flow of gas. For example, Quicker and Deckwer (1981) based their superficial gas velocity on the pressure at the bottom of their column, while in the work of Reilly et al. the superficial gas velocities are calculated based on the pressure at the midpoint of the column. Unfortunately most investigators fail to mention the conditions on which they based their superficial gas velocity calculations.

Since most investigators work at atmospheric conditions quite a significant change in superficial gas velocity can result depending on column (liquid) height and choice of reference point. For example, assume that the superficial gas velocities are calculated for a 0.15 m column with a constant mass flow of air, such that at atmospheric pressure this flow results in a superficial gas velocity of 0.10 m/s. If the column was 5 m high and contained an air/water mixture, then the superficial gas velocity based at the column bottom (p = 15 MPa) would be 0.0667 m/s. If the reference point were taken halfway up the column, as in this work, the superficial gas velocity would be 0.0804 m/s, a difference of 20%.

In systems in which the bubbles are not at their maximum equilibrium size and in which some coalescence occurs, the liquid height will influence the extent of coalescence. For example, Wallis (1969) and Voigt and Schugerl (1979) using shallow systems found the gas holdup to decrease with liquid height, because the longer the column the more time the bubbles have to coalesce. Other investigators report no influence of the liquid height [Hikita and Kikukawa (1974), Deckwer et al. (1980), Godbole et al. (1984), for height to diameter ratios, H/D, ranging from 5.74 to 9.91] while Patil et al. (1984) observed only a very small influence of the liquid height when varying H/D from 1.8 to 2.7. Similarly, Koide et al. (1984) reported that the influence of the static slurry height became very small for heights greater than 1 m. Higher H/D ratios did not result in a further decrease of the gas holdup. Observed variations appear to be more a function of the system than the H/D ratio. It would appear that gas holdup is independent of H/D rates for columns where H/D is >5. Higher H/D ratios did not result in a further decrease of the gas holdup.

LIQUID FLOW RATE: Several investigators state that the liquid velocity does not affect the gas holdup [Voigt and Schugerl (1979), Akita and Yoshida (1973), Shah et al. (1982)]. However, Kara et al. (1982) observed

that the gas holdup decreased with increasing liquid velocity at higher gas rates, as was also found by Hills (1976), Pal et al. (1980), Buchholz et al. (1983) and Kelkar et al. (1984). Kelkar et al. (1984) measured the gas holdup in the presence of solids and found that the superficial slurry velocity had the greatest effect when the gas and slurry rates were comparable, i.e., at low gas rates. It appears that the effect of the liquid flow rate is at most small, and for the relatively low flow rates usually employed in bubble column reactors, can probably be ignored. It is only at higher liquid rates that the effect appears to merit consideration.

GAS DENSITY: Summaries of correlations such as those given by Shah et al. (1982) indicate most investigators consider that the gas density has little or no effect on the gas holdup. However, Akita and Yoshida (1973) found a slightly lower gas holdup when using helium. Bhaga et al. (1971) noted that denser gases resulted in higher gas holdups. Hikita et al. (1980) also observed some effect of gas density. Finally, Mersmann (1978) in his semi-theoretical derivation of correlations describing gas holdup and bubble size incorporated a slight dependence on the gas density.

PRESENCE OF SOLIDS: Various studies agree that solids reduce the gas holdup slightly [Deckwer et al. (1980), Linneweber and Blass (1983), Ying et al. (1981), Kara et al. (1982), Kelkar et al. (1984), Koide et al. (1984)]. Solids concentrations investigated were up to 16 wt %, up to 9-10 wt %, up to 45 wt %, 0-40 wt %, up to 15 wt % respectively. Quicker and Deckwer (1981) found a significant decrease in holdup, the decrease being larger with increased solids loading. Ying et al. (1981) found that increasing the solids concentration from 15.7 to 45 wt % did not reduce the gas holdup any further. Kara et al. (1982) reported that the gas holdup decreased with increasing solids concentration, but that the effect became less at higher solids concentrations and higher gas rates. They allowed for the effect of the solids on gas holdup by using the slurry viscosity instead of liquid viscosity in their proposed correlation. Quicker and Deckwer (1981) tried to describe the gas-liquid interfacial area as a function of U_G and μ_{eff} but found that the effective viscosity as determined in a concentric cylinder viscometer provided insufficient characterization. Contrary to Kelkar et al. (1984) they reported that an increase in particle size reduced the gas holdup. Kelkar et al. (1984) noticed an effect of the particle wetability on the gas holdup; hydrophilic particles increased the bubble coalescence [See also Bhatia et al.

(1972)]. Koide et al. (1984) found the effect of solids on gas holdup to be larger in the transition region from bubbly to churn-turbulent flow than in the churn-turbulent region.

COMPARISON OF CORRELATIONS: As an example of the differences in gas holdup predictions of existing correlations, four general correlations were selected and are given in Table 1 [Akita and Yoshida (1973), Bach and Pilhofer (1978), Hikita, Asai, Tanigawa, Segawa, and Kitao (1980), Kumar, Degaleesan, Laddha and Hoelscher (1976)].

The gas holdup predicted by these correlations was plotted versus the superficial gas velocity for three air/liquid systems: air/water, air/trichloroethylene and air/Varsol (Varsol DX 3641 is a light hydrocarbon oil available from ESSO Chemicals). The physical properties of the liquids are listed in Table 2.

Figures 1 to 3 show the predictions of the four correlations for each of the systems. Due to the type of expression used by Kumar et al. maxima and minima (at higher gas velocities) result. Apparently this expression should only be used for gas rates below 0.10 m/s. Fairly large differences result between the different correlations, even at low gas rates. The Akita and Yoshida and the Hikita et al. correlations compare very well for the air/water and air/trichloroethylene and a little less well for the air/Varsol system. In both these studies single spargers were used and the holdup was measured over a wide range of gas rates (0-0.40 m/s and 0.042-0.38 m/s respectively). Bach and Pilhofer measured holdup at gas velocities between 0.01-0.20 m/s and used a perforated plate distributor. Kumar et al. measured between 0.02-0.1 m/s for the air/water system and at lower gas rates for other liquids. A perforated plate was used and most experiments were done in a 0.05 m diameter column; only a few experiments with air/water and air/NaOH solutions were performed in 0.075 and 0.10 m columns.

Some of the differences in the predictions might be the result of the difference in the ranges of gas rates that were used by the different authors. Experiments mainly measuring holdup in the bubble flow regime and in the transition regime to churn-turbulent flow obtained the highest n value ($\varepsilon_G \propto U_G^n$). At higher gas rates these correlations would be expected to predict holdup values that are too high. Another factor which may possibly account for some of the differences in predicted gas holdup values is the difference

in spargers used (perforated plate versus single orifice). As mentioned above, different spargers and different sparger openings can result in widely different gas holdups at lower superficial gas velocities. However, the differences in the gas holdup predictions between the four supposedly fairly general holdup correlations, suggest that they lack general applicability. At higher gas flow rates in the turbulent regions where effects due to sparger design or limited coalescence should be minimal, they fail to provide an adequate prediction of holdup in systems other than those used in their development, for example, predictions of holdup for liquids such as Varsol or trichloroethylene show substantial disagreement with experiment.

NOMENCLATURE

d_B	bubble diameter, cm
d_o	orifice diameter, m
$\bar{d}_p$	mean particle diameter, μm
D	column diameter, m
g	gravitational constant, m/s^2
H	static liquid height, m
N_{Re_o}	single particle Reynolds number for settling
M	number of orifices
P/M	power input/unit mass m^2/s^3
U_G	superficial gas velocity, m/s
$\bar{U}_G$	column averaged superficial gas velocity, m/s
V_o	single particle free settling velocity, m/s
ϵ_G	gas holdup, volume fraction
ρ_G	gas density, kg/m^3
ρ_L	liquid density, kg/m^3
σ	surface tension, kg/s^2
ν_L	liquid kinematic viscosity, m^2/s
μ_G	gas viscosity, kg/ms
μ_L	liquid viscosity, kg/ms

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Table 1 - Selected gas holdup correlations [Shah et al. (1982)]

System	Range of parameters	Correlation	References
1. Air-water Air-glycol aq. soln. Air-methanol O_2-water	U_G, m/s 0.003 - 0.4 U_L, m/s 0 - 0.044 D, m 0.152 - 0.6 H, m 1.26 - 3.5 Single nozzle sparger d_o, m, 0.005	$\frac{\epsilon_G}{(1-\epsilon_G)^4} = C\left[\frac{g D^2 \rho_L}{\delta}\right]^{1/8} \cdot \left[\frac{g D^3}{\nu_L}\right]^{1/12} \cdot \left[\frac{U_G}{\sqrt{g D}}\right]$	Akita and Yoshida (1973)
2. Air-alcohols Air-halogenated hydrocarbons	U_G, m/s 0 - 0.2 D, m 0.1 H, m > 1.2 Perforated plate d_o, m 0.0005, 0.001	$\frac{\epsilon_G}{1-\epsilon_g} = 0.115\left[\frac{U_g^3}{\nu_L g(\rho_L-\rho_G)/\rho_L}\right]^{0.23}$	Bach and Pilhofer (1978)
3. Air-water Air-glycerol aq. soln. Air-kerosene	U_G, m/s 0.0014 - 0.14 D, m 0.05, 0.07, 0.10 Perforated plate d_o, m 0.00087 - 0.00309 M = 21-49	$\epsilon_G = 0.728\,U - 0.485\,U^2 + 0.0975\,U^3$ $U = U_G\left[\rho_L^2/\sigma(\rho_L-\rho_G)g\right]^{1/4}$	Kumar et al. (1976)
4. (Air, H_2, CO_2, CH_4, C_3H_8, N_2) - water Air-organic liquids Air-electrolyte soln.	U_G, m/s 0.012 - 0.38 D, m 0.1 H, m 0.65 Single nozzle sparger d_i, m 0.011	$\epsilon_G = 0.672\,f\left[\frac{U_G\mu_L}{\sigma}\right]^{0.578}\left[\frac{\mu_L^4 g}{\rho_L\sigma^3}\right]^{-0.131}\left[\frac{\rho_G}{\rho_L}\right]^{0.062}\left[\frac{\mu_G}{\mu_L}\right]^{0.107}$ f = 1.0 for non-electrolytes	Hikita et al. (1980)

Table 2 - Properties of liquids used in 0.30 m diameter bubble column

	Water*		Varsol**		Trichloroethylene***	
	25°C	40°C	25°C	35°C	15°C	25°C
ρ_L, Liquid density (kg/m^3)	997	992	788	780	1468	1452
μ_L, Liquid viscosity ($Pa.s \times 10^3$)	0.890	0.653	1.236	1.052	0.609	0.552
P_{VAP}, Vapour pressure (kPa)	3.17	7.37	0.5 @ 38°C		6.00	10.00
σ_L, Surface tension (N/m)	0.0720	0.0696	0.0283 @ 25°C		0.029 @ 30°C	

*Deionized

**Esso Chemicals Varsol Solvent DX 3641, low aromatics, 185°-209°C B.P.

***Canada Colours and Chemical Ltd., Neu-Trichloroethylene (Dow Chemical Co.)

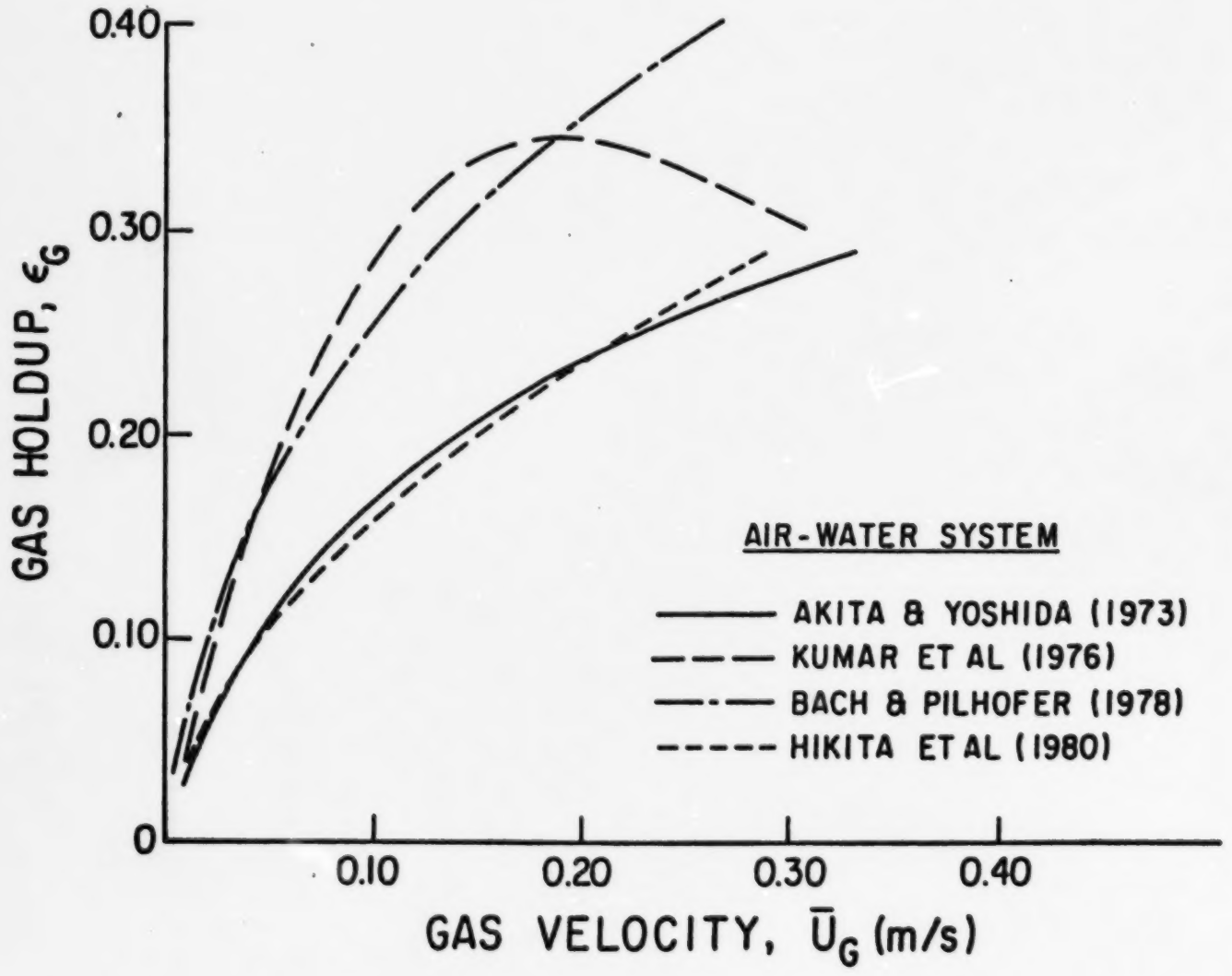

Fig. 1 - Comparison of gas holdup prediction for four typical holdup correlations, air-water system, 20°C, 1 atm

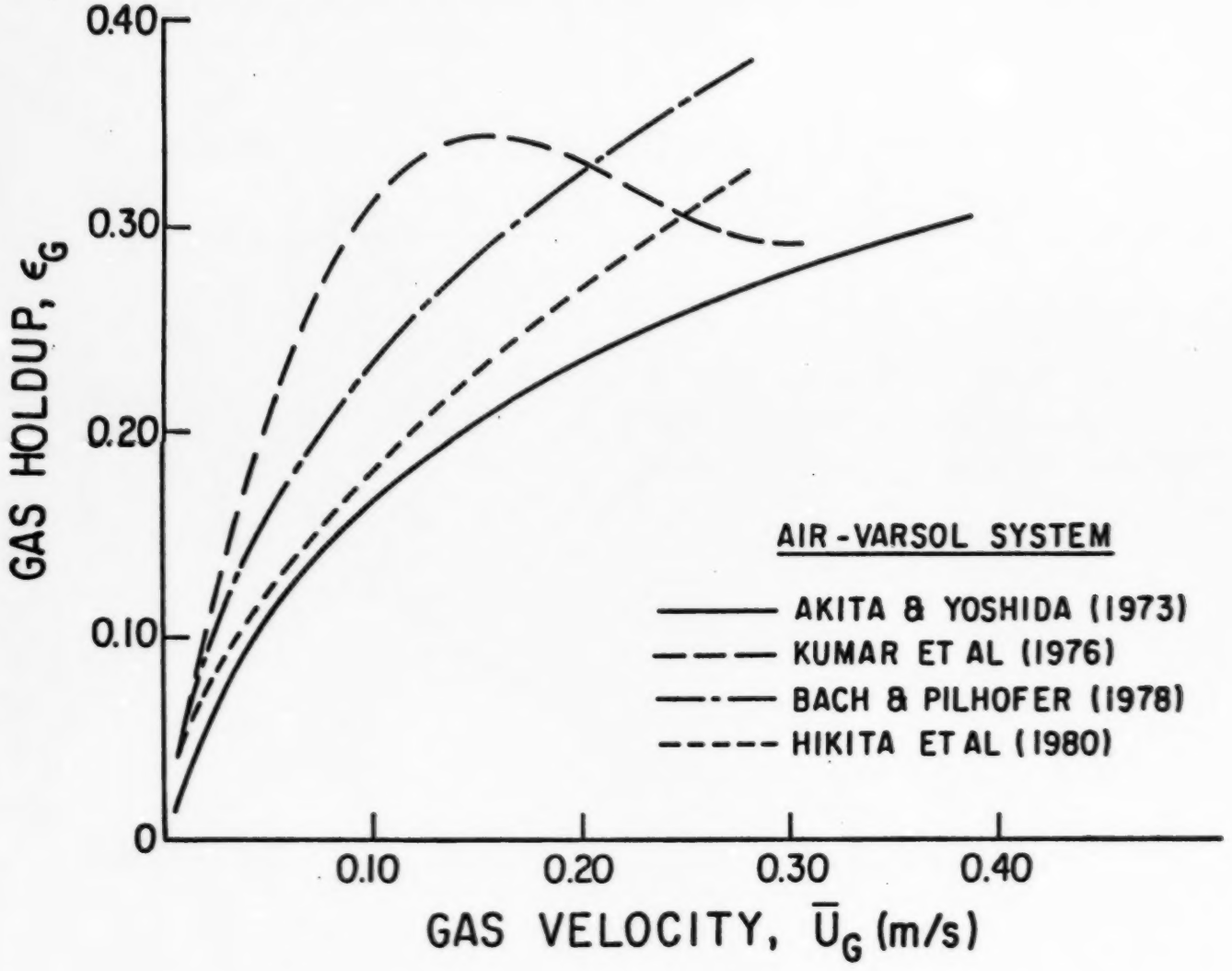

Fig. 2 - Comparison of gas holdup prediction for four typical holdup correlations, air-varsol system, 20°C, 1 atm

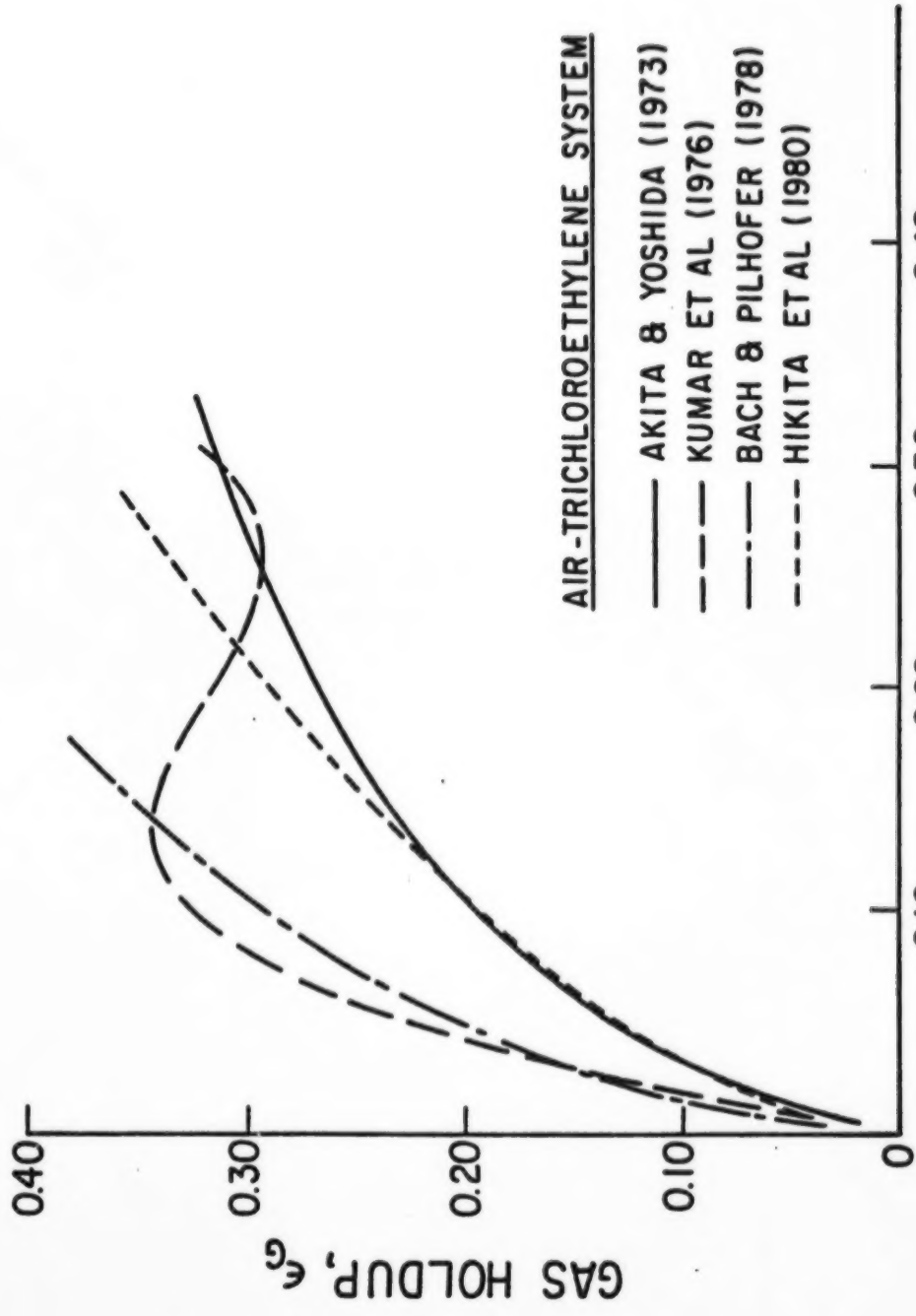

Fig. 3 - Comparison of gas holdup prediction for four typical holdup correlations, air-trichloroethylene system, 20°C, 1 atm